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Spin-Precession Organic Magnetic Sensor

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EXECUTIVE SUMMARY

This report documents the results of $CoFe_{50}Al_{25}Si_{25}$ (CFAS) studies carried out by SRI International in collaboration with Professor Nate Newman's group at Arizona State University (ASU) under ONR Contract N00014-09-C-0292. In this period we have: (1) grown and characterized—both electrically and magnetically— a new half-metal CFAS that has desirable properties for use at room temperature; (2) fabricated several nonlocal devices with CFAS and polymer for magnetic sensing; and (3) studied the performance of these devices for a range of magnetic fields and temperatures.

Our earlier studies using a different half-metal LaSrMnO (LSMO) and polymer (P3HT) were able to demonstrate magnetic sensor operation at low temperature. Spin injection, spin precession, and spin detection requirements for ultra-high sensitivity were observed only at low temperatures. Since the Curie temperature of the LSMO is only slightly higher than room temperature, the magnetization at room temperature is very small. This prevented efficient spin injection, resulting in a marked drop in the signal amplitude and a signal-to-noise ratio (SNR) too small for useful sensor performance.

CFAS is a half-metal with a high Curie temperature (T_C ~1000 K). High-quality CFAS thin films produced using pulsed laser deposition and DC magnetron sputtering exhibit very high and temperature-*independent* magnetization (~ 1000 emu/cc), high coercive field (~ 450 Oe), low resistivity (~50 μΩ-cm), and large spin polarization ($p > 0.6$) at room temperature. We have fabricated nonlocal devices using these films with the goal of producing useful room temperature sensors. Preliminary results show the possibility of sensitive room temperature operation. Device optimization is in progress. This report summarizes the results of our studies to date. A comprehensive review and further results will be presented at the annual review in August 2012.

TECHNICAL DISCUSSION

1. Objectives

Our long-term goal is to develop an ultrasensitive, room temperature, compact magnetic sensor based on spin precession. The specific aim of this study is to fabricate a magnetic sensor with half-metal ferromagnets (FM) as contacts and polymer as a transport medium to demonstrate and optimize the sensing capability of the device. The sensitivity depends crucially on efficiency of spin injection and duration of spin relaxation time. The half-metallicity ensures high spin injection, and the small spin-orbit interaction in polymers ensures long spin lifetimes. We consider $CoFe_{50}Al_{25}Si_{25}$ (CFAS) as possible candidates for half-metallic FM contact and poly(3,4-ethylenedioxythiophene) (PEDOT), poly-3(hexylthiophene) (P3HT), or poly[2-methoxy, 5-(2-ethylhexoxy)-1,4-phenylene vinylene] (MEH-PPV) as candidates for polymers.

2. Device Concept, Operation, and Approach

The device considered here is a planar structure that uses a half-metal such as LSMO or CFAS for the contacts and an organic material for electron transport. The contact “in-plane bar magnets” are magnetically poled to have magnetization parallel to each other. The half-metal nature of the contact allows injection of only one kind of spin (parallel to the magnetization) into the organic material. The constituents of the organics are usually atoms with small atomic numbers (such as H, C, N, O, and S), and the spin-orbit coupling is extremely small, which allows the injected spins to remain coherent for long periods of time. In the absence of any magnetic field, the injected electrons will retain their spin polarization and can find a state in the other contact when the two contact magnetizations are parallel. However, when a magnetic field is applied, the electron spin precesses and the spin orientation with respect to contact magnetization changes, resulting in increased resistance. Since a very small magnetic field is required to alter the electron spin orientation, the device is predicted to have very high sensitivity, even at room temperature.

3. Materials Synthesis and Characterization

Previously we considered local devices with LSMO as contacts. The spin injection was demonstrated at low temperature (35 K). However, because of LSMO’s low Curie temperature, local devices did not show room temperature spin injection. To overcome this issue, we have focused our efforts on the growth, characterization, and optimization of CFAS, a half-metal material system with a ~1000 K Curie temperature. Pulsed laser deposition (PLD) and magnetron sputtering methods were used to deposit CFAS thin films. We considered two approaches to improve materials quality: high-temperature growth in a one-step process and room temperature growth followed by a high-temperature anneal in a two-step process.

The magnetic hysteresis curve obtained for a sample grown by the one-step process is shown in Figure 1. Similar results are obtained also for two-step growth samples. Note that both the coercive field (H_c) and magnetization (M_s) of the CFAS films are large, thus boding well for their use in a magnetic sensor operating at room temperature. The results of our systematic study of the H_c and M_s as a function of growth temperature (for the one-step method) and anneal temperature (for the two-step method) are summarized in Figure 2.

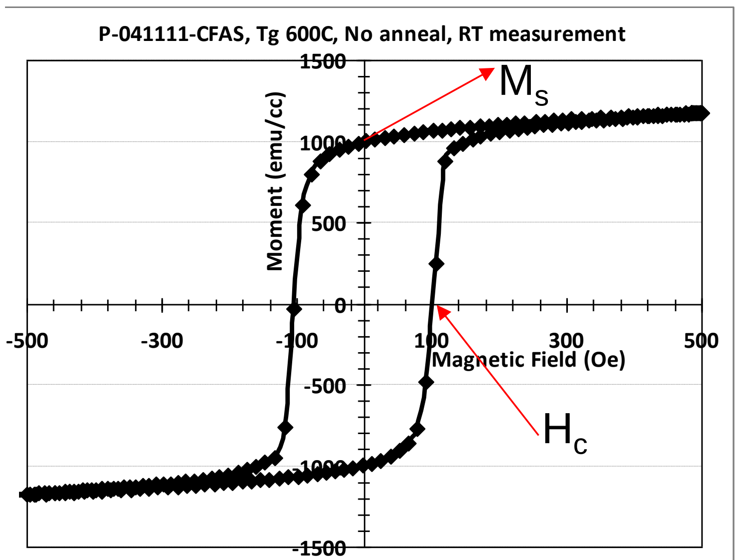

Figure 1 Measured magnetization of CFAS as a function of applied field.

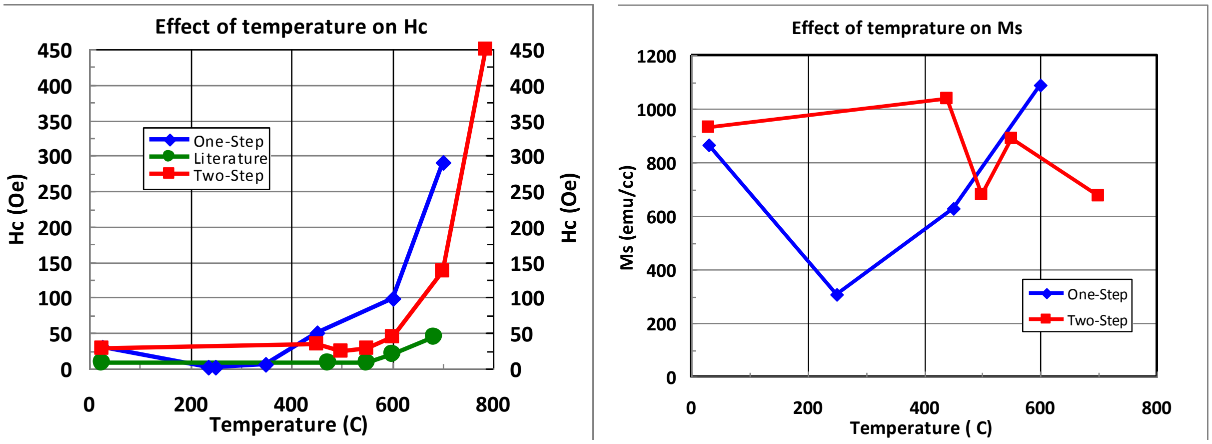

Figure 2: Magnetic properties of CFAS layers measured at room temperature as a function of growth (one-step) or anneal (two-step) temperature. Also shown (green) is best reported literature value.

H_c monotonically increases with the growth temperature in the one-step process, with the optimized values significantly larger than the best published CFAS values (green line in Figure 2). For the one-step process, M_s is larger only when higher substrate temperatures are used, where it is nearly temperature-independent for the two-step process.

We also measured the transport properties of CFAS layers. The electrical resistivity is small (~60 μΩ-cm) and changes very little with temperature. The low resistivity confirms its metallic state, and the temperature independence suggests that strong alloy scattering is present. The magnetoresistance is also very small (~ 140 μΩ-cm) and is nearly independent of the magnetic field (about 0.1 Ω change when exposed to a 3T magnetic field.

We have further measured the degree of spin polarization, *p*, at the CFAS surface at room temperature using the Andreev reflection technique. Initial work measured a large spin polarization value of 0.6. We are currently performing a systematic study to optimize the amount of spin polarization at the surface as a function of growth conditions. The results will be reported in the annual review to be held in August 2012.

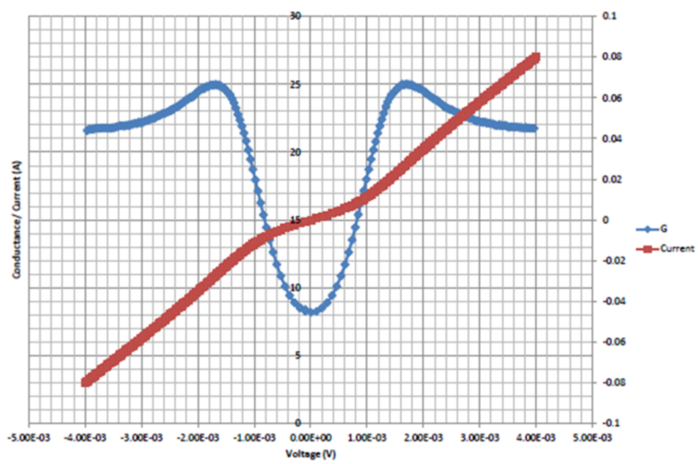

Figure 3: Current–voltage (red) and conductance-voltage (blue) for a permalloy (bottom electrode)-native oxide (tunnel barrier)-Pb (superconducting top electrode) demonstrating the dominance of tunneling. Note the peak near the Pb's energy gap of 1.4 meV. This method will be used to optimize the tunnel barrier at the CFAS surface in future studies.

To achieve efficient spin injection from the half-metal electrode, a tunnel barrier must be produced at the CFAS surface. We have begun a study that can determine the fraction of tunneling current injected from a magnetic surface by constructing a structure of a magnetic permalloy bottom electrode (NiFe), exposing it to air to form a native oxide, and then depositing a Pb superconductor layer. The presence of an energy gap in the superconducting electrode allows us to discern tunneling current from all others. If the magnitude of current that flows below the gap voltage of the Pb superconductor (i.e., ~1.3 meV) is from thermal pair breaking alone and not from pinholes or transport over the barrier, we can conclude that all of the current is from electron (quasi-particle) tunneling. If tunneling dominates, spin injection will be very efficient. Figure 3 shows that this is in fact the case for the permalloy native oxide tunnel barrier. Work is under way to use this method to detect the fraction of tunneling current flowing from CFAS structures with native oxides and deposited AlO_x barriers.

4. Nonlocal CFAS Devices Fabrication and Results

The Curie temperature of our CFAS material is estimated to be in excess of 1000 K, and magnetization is shown to be relatively independent of temperature between 4 K and 300 K. All these features are expected to increase the performance and stability of the magnetic sensor. For noise considerations, we chose to focus our efforts on a nonlocal device, as shown schematically in Figure 4. The terminals 1 (T1) through 4 (T4) are CFAS lines (in the z direction) on an STO substrate with carefully chosen widths (w_i) and separated by selected distances (d_{ij}). The polymer is drop-cast on CFAS lines. The CFAS contacts are magnetized. A constant current is supplied between T1 and T2. Depending on the magnetization of T2, charge carrier spins (parallel to the magnetization) accumulate under T2 and diffuse toward T3 and T4. In a steady-state distribution of spins, the potential measured between T3 and T4 depends on the density of carriers with spins parallel to their respective contact magnetization at T3 and T4. Since this is an open circuit voltage, no charge transport between T3 and T4 takes place. Consequently, carrier recombination, trapping, and other issues at interfaces no longer contribute to noise. This material combined with this design thus potentially increases the signal and decreases noise, and is therefore likely to enable high-sensitivity room temperature operation.

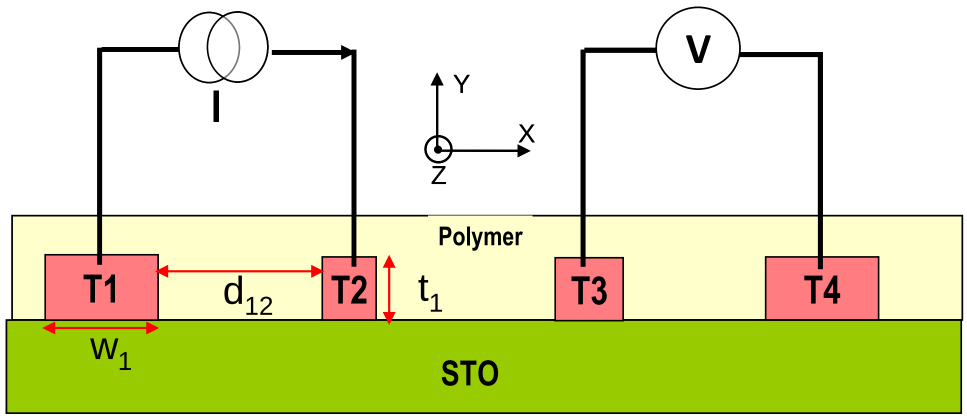

Figure 4: Schematic nonlocal CFAS device.

To date we have fabricated several nonlocal devices with device typical dimensions— $w_1 = w_4 > 2$ µm, $w_2 = 0.5$ µm, $w_3 = 1.0$ µm, $d_{12} = d_{23} = d_{34} = 1$ µm, and $t_1 = t_2 = t_3 = t_4 > 30$ nm—in the architecture shown in Figure 5 (left). The length of the CFAS lines is 200 µm. In our initial work, the polymer droplets, deposited manually by the drop-cast method, were larger than 200 µm and partially shorted the current between the Au pads. In these devices, a measurable spin injection was not observed. We have subsequently used a state-of-the-art polymer deposition system—Rain Maker—developed by ASU for commercial use to deposit precisely sized polymer droplets far smaller than 200 µm, as seen in Figure 5 (right). This allows the polymer droplet size to be small enough to avoid contact with the Au pads, maximizing the spin-polarized current injected by CFAS. We have fabricated several devices and measured their magnetic response. A constant current is applied between terminals 1 and 2, and the voltage is measured between terminals 3 and 4 as a function of applied magnetic field.

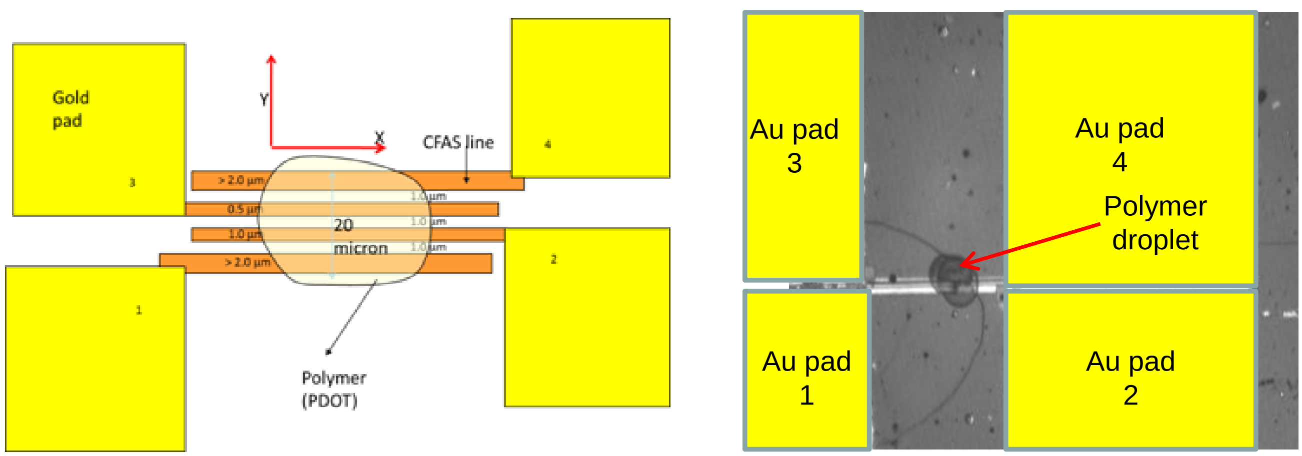

Figure 5: Nonlocal device architecture (left) and the fabricated device (right) with PEDOT deposited by Rain Maker.

The nonlocal resistance (ratio of voltage to current) is plotted in Figure 6 as a function of magnetic field applied parallel to the CFAS strips. When the magnetization of the terminal 2 "bar magnet" is reversed, the voltage measured between terminals 3 and 4 will change sign, resulting in a change in sign of the non-local resistance (V_3-V_4/$I_{1\text{-}2}$). This behavior is clearly seen near 50 Oe for one of the fabricated devices (Figure 6). As the current is increased from 60 pA to 500 pA, the relative noise in the signal V_3-V_4 voltage increases. Efforts are under way to confirm proper device operation. Future work will involve the optimization of the device's magnetic field sensitivity by altering the material growth, processing and microfabrication conditions and the device dimensions and operating parameters.

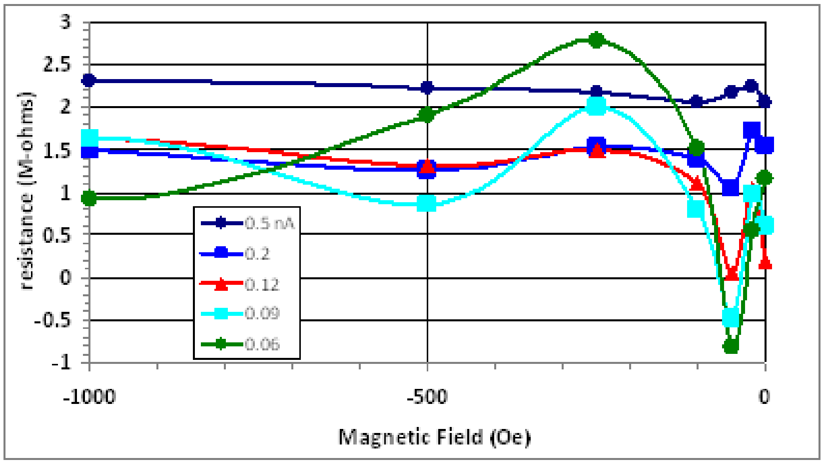

Figure 6: Measured magnetic response of CFAS non-local device

5. Conclusions

In conclusion, we have synthesized and characterized CFAS thin films, which consistently exhibit excellent electrical and magnetic properties. We have fabricated several nonlocal devices and measured their magnetic response. To date, the devices have very small SNR. We are currently working to independently optimize the spin injection efficiency and reduce the noise sources. We will then systematically adjust the relevant parameters to optimize device performance. The results of our systematic study will be summarized and reported at the ONR annual review to be held in August 2012.